

## Piezocomposite of Fine PZT Rods Realized with Synchrotron Radiation Lithography

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### Abstract

A ceramics micro fabrication technique has been developed for the 1-3 piezocomposite which is suitable for high frequency, wideband ultrasound transducers. This process employs synchrotron radiation (SR) lithography and a micro molding process, generally called the "LIGA process". This process realized an array of lead zirconate titanate (PZT) rods whose cross section is 25 micron. The composite made up of circular sectional PZT rods was clarified not to have an internal lateral frequency mode and it is available over a wide frequency range. The piezocomposite was clarified as having predicted merits, and was thought to contribute to the improvement of the resolution.

### Introduction

Medical ultrasonic diagnosis has been widely used because of its weak influence on patients, ability of blood flow measurement, and real time measurements. The improvement of the resolution is always required, therefore many techniques are being studied. To achieve this, in particular, the changing of the transducer material from piezoelectric ceramics like PZT to 1-3 piezocomposites as shown in Fig. 1 is a thorough idea [1,2].

The composites are expected to be suitable materials to improve the resolution because of their low mechanical quality factor ( $Q_m$ ) and low acoustic impedance ( $Z$ ). However, by the conventional dice-and-fill method, it is impossible to fabricate PZT rods which are small

enough to make the composites behave like homogeneous materials. The size of the PZT rods is required to be 25 micron, which would allow the operating frequency to be expanded above 10 MHz [3]. New techniques, such as the fugitive mold technique [4] and a injection molding process [3], have been developed, however they are not sufficient.

We improved the fugitive mold technique and formed an array of PZT rods of diameters less than 20 micron and an aspect ratio of over 5 [5]. As a result, piezocomposites for high frequency ultrasonic probes were made available. The effects of the Young's modulus of the polymer, the volume fraction of PZT and the aspect ratio of the PZT rods on the electromechanical coupling coefficients have been reported previously [6,7].

However, this method requires a new plastic mold formed by SR lithography for every composite, therefore a limitation exists in terms of cost. We developed a mass-production process and succeeded in expanding the size of the composites. This makes piezocomposites for high frequencies industrially available for the first time.

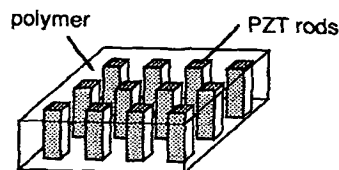


Fig.1 Schematic view of piezocomposite.

### Fabrication process

The fabrication process is shown in Fig.2.

Using SR lithography, a resist structure with deep holes is formed. Then, electroforming is performed and the complementary nickel mold insert is made. By molding with this, acrylic molds are fabricated economically. This process is generally called the "LIGA process" [8].

After a PZT slurry is injected into an acrylic mold, this mold is removed by plasma etching [5]. In the fugitive mold technique, a burn-out process is usually used to remove the plastic mold [4]. However the fine PZT rods with a high aspect ratio, topple during this process. Sintering is performed and a PZT rod array is obtained. The array is then cast in epoxy resin by vacuum impregnation. The remaining epoxy resin and PZT base are polished until the rod surfaces appear. Electrodes are sputtered and poling is performed. The resist structure, the mold insert, the plastic mold and the PZT array are shown in Fig.3.

The LIGA process was originally developed in the early 1980s in Germany, and has become one of the most expected micro-fabrication processes. However the mold insert with such a high aspect ratio and fine rods as shown in Fig.3 has not been realized till now. It was realized by optimizing the electroforming condition and the pretreatment by ultrasound, and

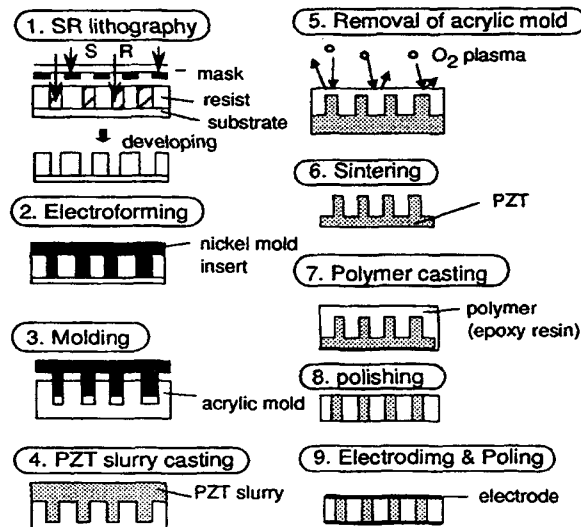
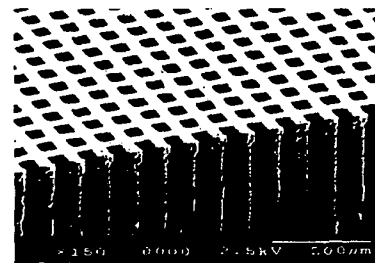
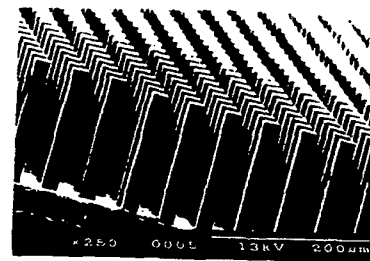


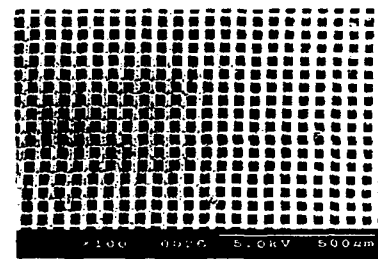
Fig.2 Brief fabrication process of piezocomposite.



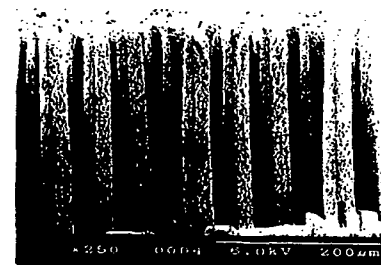
(a) Resist structure.



(b) Nickel mold insert.



(c) Acrylic mold.



(d) As-sintered PZT rods array.

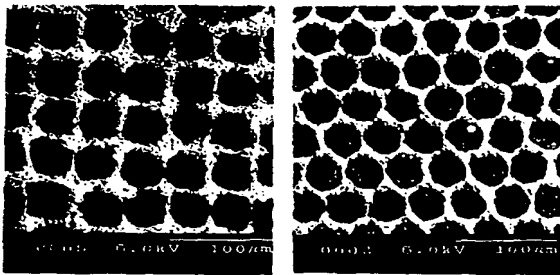
Fig.3 SEM photographs. The sectional size of PZT rods is 25 micron and the height of them is 250 micron.

developing an effective method to stir the solution into the deep and fine holes [9]. The micro molding is also difficult because of the large surface area, however an effective internal mold release agent made it possible.

To expand the size, it was necessary to control the bending of the PZT base while sintering. If the bending is excessive, a small area is available after the polishing because the height of the PZT rods is limited. Annealing at 1300 centigrade is effective to suppress the bending under 50 micron /15 mm, and the size of composite was larger than 15x10 mm<sup>2</sup>, when the thickness was 120 micron.

The electrode is composed a 0.1 micron-thick chromium layer and a 0.4 micron-thick gold layer. The sticking force is measured by the peel test and clarified as beyond 300 g/mm<sup>2</sup>. It is enough for the electrode.

This process enables the tailoring of the piezocomposites suitable for applications by a free choice of the sectional shape, the volume fraction and the distribution of the PZT rods. Only the mask design needs to be changed to change the geometry of the PZT. Fig.4 shows the surface of the different geometry composites. Sample parameters are given in Table I.



(a) Square section.

(b) Circular section.

Fig.4 Geometry of PZT rods (volume fraction; 40%). Black parts are PZT.

Table I. Parameters of fabricated piezocomposites.

size of PZT	25 micron
section of PZT	square, circle
volume fraction of PZT	25%, 40%
aspect ratio of PZT	2.5- 7

## Properties

### Internal Lateral Resonance

The available frequency range of the developed composite was investigated.

There is a microstructure in the piezocomposites, therefore many vibration modes exist which are not seen in homogeneous materials. When the acoustic waves traveling in the plane, the waves are reflected in the lateral periodicity of the composites' microstructures and lateral mode resonances are set up. If such unwanted resonances occur in the operating frequency band of the transducer, the desired thickness mode resonance is corrupted. The internal lateral resonance should be pushed above twice the operation frequency, therefore the maximum operation frequency is half the internal lateral resonance frequencies [10].

The internal lateral mode resonances were measured for the samples are given in Table I.

In the square section geometry, lateral mode resonance appears as shown in Fig.5(a). When the volume fraction of PZT is 25 % and 40 %, despite the aspect ratio of the PZT rods, resonance existed at 14-15 MHz and 20-21 MHz, respectively. Their respective electromechanical coupling coefficients ( $k$ ) were about 25 % and 20 %. The pitch of the rods was 50 micron and 38 micron, respectively, and the velocity in the plane was measured and approximately 1500 m/s. Therefore, it is reasonable to recognize the resonances as the internal lateral mode. Therefore these composites should be used beneath 7.5 MHz and 10 MHz, respectively. Finer pitch would be required for operation over these frequencies.

However, in the circular section geometry, lateral mode resonances were weak and not detectable as shown in Fig.5(b). The reason for this is thought to be as follows; the nodes form a line in square section geometry and the same periodicity exists in many places, however the nodes are scattered in the plane in circular geometry and the same periodicity exists in a few places.

Therefore, the piezocomposite of the circular section geometry is clarified as not to

have an upper limit of operating frequency and to be available over a wide frequency range.

### Piezoelectric and Dielectric Properties

The properties of the composites which are composed of circular PZT rods with 40 % volume fraction are shown in Fig.6. In this figure, the thickness of the composites expand from 60 micron (aspect ratio: 2.5) to 180 micron (aspect ratio: 7). The resonant frequencies of the thickness mode is 23 MHz and 7.5 MHz, respectively.

$Q_m$  was calculated by the admittance circle around the resonant frequency of the thickness mode. These values increased as the aspect ratio increased, however they were very low in comparison with the value of 27 for monolithic PZT. Therefore the shortening of pulses is possible, and the axial resolution is expected to be improved.

$k$  was determined by impedance measurements and equivalent circuit fitting. In the thickness mode,  $k_t$  increased as the aspect ratio increased. Even if the aspect ratio was 2.5,  $k_t$  of the composite was 60 %. It was larger than the  $k_t$

of 47 % for PZT. Therefore, the S/N ratio would be larger. In the width mode,  $k_{31}'$  also increased as the aspect ratio increased. Even if the aspect ratio was 7,  $k_{31}'$  was 20 % and much lower than the  $k_{31}'$  of 45 % for PZT. This is thought to be effective to decrease the crosstalk.

$Z$  is defined as the product of the density and acoustic velocity ( $c_p$ ).  $c_p$  was calculated from  $c_p = 2N_t$ .  $N_t$  was a frequency constant in the thickness mode and was calculated as the product of the antiresonance frequency and thickness.  $N_t$  was approximately 1800 kHz\*mm for all cases of this measurement. Therefore, the estimated  $Z$  was 13.4 MRayl. This value is much lower than the  $Z$  of 30 MRayl for monolithic PZT. The acoustic impedance of the human tissue is 1.5 MRayl, therefore the acoustic energy is transferred more effectively to the tissue. By a simple calculation, the acoustic transparencies were 20 % for the composites and 9.5 % for PZT. Therefore, the S/N ratio would be greater.

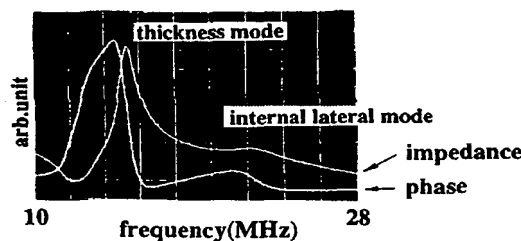
As mentioned above, the predicted merits of piezocomposites are recognized. The developed piezocomposite is thought to contribute to the improvement of the resolution.

The relative dielectric constant ( $K_{33}^T$ ) is also measured.  $K_{33}^T$  of PZT in this work is 5700. The measured  $K_{33}^T$  of the composites is 1850. This means that  $K_{33}^T$  of micro rods is 4600. It is 20% lower than that of bulk PZT. The reason is thought to be because the average grain size is about 8 micron and the effects of surface and boundary are dominant. However, 1850 is a good value of  $K_{33}^T$  for the composites, and it is easier to obtain electrical impedance matching with the cables.

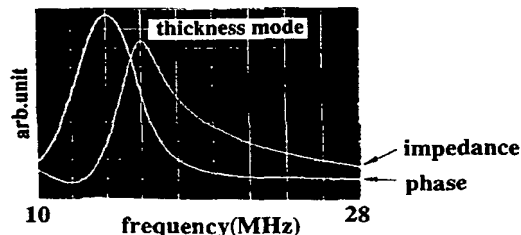
The properties are summarized in Table II associated with the applications.

### Conclusion

A ceramics micro fabrication technique, which employs the LIGA process, has been developed and the piezocomposite with 25



(a) Square section geometry (volume fraction; 40%, aspect ratio; 5)



(b) Circular section geometry (volume fraction; 40%, aspect ratio; 5)

Fig.5 Measured impedance and phase.

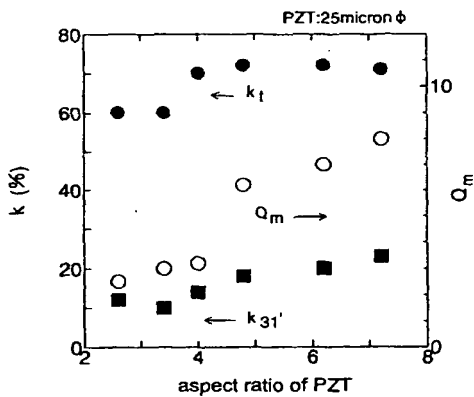


Fig.6 Dependence of  $k$  and  $Q_m$  on aspect ratio of PZT rods (volume fraction; 40%) .

Table II. Trial calculation properties associated with applications.

application	intra-vascular	high freq. echo
operation freq.(MHz)	23	7.5
element size (mm)	0.7 x 0.7	0.2 x 10
thickness (micron)	60	180
$k_t$ (%)	60	71
$k_{31}'$ (%)	12	23
impedance of element ( $\Omega$ )	52	117

micron rods were fabricated. The effects of the sectional shape of the PZT rods are investigated. The composites made up of circular sectional PZT rods were clarified not to have an internal lateral frequency mode and are available over a wide frequency range. The piezocomposite was clarified to have predicted merits; low  $Q_m$ , high  $k_t$ , low  $k_{31}'$ , and low  $Z$ . This was thought to contribute to the improvement of the resolution.

#### Acknowledgement

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# NOVEL PROCESSING OF HIGH ASPECT RATIO 1-3 STRUCTURES OF HIGH DENSITY PZT

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## ABSTRACT

A new process for the fabrication of fine scale, high aspect ratio PZT 1-3 composite structures has been developed. In this process, lost mold technique is utilized with a Si mold prepared by deep reactive ion etching (RIE). Due to the high strength and high melting point of Si, PZT sintering under high pressures without removing the mold becomes possible, leading to structures of highly condensed PZT in the precise shape of the mold. The typical PZT structures in this work are periodically arrayed rods 16  $\mu\text{m}$  in diameter, 100  $\mu\text{m}$  in height, resulting in an aspect ratio higher than six. The fabricated structures are to be applied to high resolution micro-ultrasonic transducers in the frequency range of 20 MHz for medical purpose.

## INTRODUCTION

Fine scale, high aspect ratio PZT/polymer 1-3 composite is a promising for high resolution micro-ultrasonic transducers for medical purposes. [1, 2] The 1-3 composite consists of an active piezoceramic PZT (lead zirconate titanate:  $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ ) rod array embedded in a passive polymer matrix, as schematically shown in Fig. 1. The composite device is advantageous because of its large electromechanical coupling and lowered acoustic impedance which matches human tissue better and leads to high resolution. Its piezoelectric property, however, is kept close to that of bulk PZT when the volume fraction is proper. [3] For high resolution,

high aspect ratio is also important.

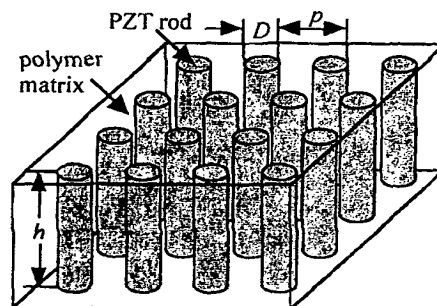


Fig. 1 Schematic of PZT/polymer 1-3 composite.

The lost mold technique is up to now most suitable for producing fine scale high aspect ratio 1-3 composite structures. [2, 4] However, in case that the mold is as fine as 10  $\mu\text{m}$  in diameter and as deep as 100  $\mu\text{m}$  in depth (an aspect ratio of 10), it is found difficult to cast PZT slurry completely into the mold because of the existence of residual air in the mold and surface tension of the slurry, even if methods such as vacuum impregnation and ultrasonic agitation are employed. Meanwhile, it is difficult to condense the molded material. As a fact, for a PZT/polymer 1-3 composite transducer, high PZT density is essential in order to obtain good transducer performance. These problems drove us to the idea of sintering PZT under high pressures. The key point is then to fabricate a mold that can withstand temperatures as high as  $\sim 1000^\circ\text{C}$  and pressures as high as tens of MPa. Obviously, the mold shall not react (or inter-diffuse) with PZT during the sintering process and shall be removable afterwards.

Previously, LIGA (lithography, galvanofarming, and plastic molding) technique [5] has been employed for the PZT 1-3 structure mold fabrication [2, 6]. By LIGA, a plastic or metal mold can be obtained. However, in the case of plastic mold, pressing at high temperatures is impossible; while a metal mold seems to be not suitable because it may react with PZT at high temperatures and may be difficult to be selectively removed.

In this paper, a new process for the fabrication of fine scale, high aspect ratio 1-3 composite structures of high density PZT is reported. In this process, a Si mold is utilized and PZT sintering is undertaken at high temperatures and under high pressures without removing the mold. The Si mold is then removed by  $\text{XeF}_2$  etching following the PZT sintering.

### TRANSDUCER DESIGNING

The underdeveloped ultrasonic transducer is designed so that it can be settled at the head of an active catheter [7], working as an imaging device. For example, it may detect the shape of a blood vessel and then guide the catheter to its destination, which is conventionally a work of X-rays. For this purpose, the transducer is in a donut geometry with its outside diameter  $D_o = 3 \text{ mm}$  and interior diameter  $D_i = 2 \text{ mm}$ , as illustrated in Fig. 2 (cross section view). A medical tool is allowed to pass through the transducer bore if necessary.

In order to obtain high resolution in a wide frequency band, the transducer is designed to work in the frequency range of 20 MHz. Additionally, in order to gain maximum sensitivity, the 1-3 composite transducer is to be operated in its halfthickness resonant mode. Therefore, the height ( $h$ ) of the PZT rods is designed as  $h \sim 100 \mu\text{m}$ . For a high resolution device, it is also essential to carefully choose the parameters such as the

PZT volume ratio  $R$ , the diameter of the PZT rod  $D$ , and the periodicity of the 1-3 structures  $p$ . In Ref. 3, Yamaguchi et al have theoretically showed that, for the 0th-order mode, the 1-3 PZT/polymer composite will strongly reflect the piezoelectric characteristics of PZT rods when  $R > 10 \sim 30 \%$  and  $\beta p < 1$ , with  $\beta$  the wave number. Following Ref. 3, the PZT 1-3 structures are designed as  $D = 10 \sim 20 \mu\text{m}$  and  $p = 10 \sim 40 \mu\text{m}$ .

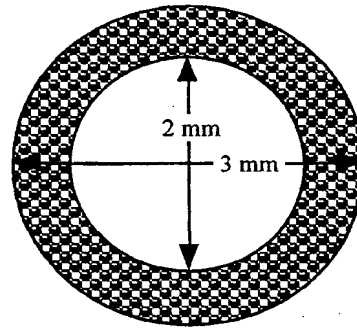


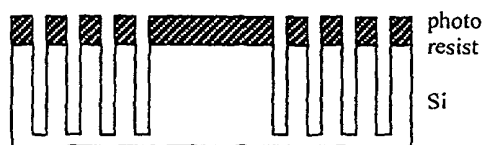
Fig. 2 Cross sectional view of the PZT/polymer 1-3 composite transducer.

### PROCESS

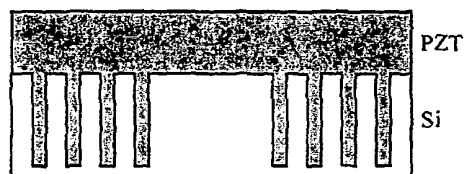
The developed PZT 1-3 structure fabrication process is shown in Fig. 3. It is a lost mold process and consists of the following steps: 1) Si mold fabrication, 2) PZT slurry casting, 3) PZT sintering under high pressures (hot isostatic pressing: HIP), and 4) Removal of Si mold.

#### 1) Si Mold Fabrication

The Si mold was fabricated by deep RIE (reactive ion etching) technique [8] (Fig. 3 a) by using an STS ICP RIE equipment. A positive photoresist, PMER P-AR 900, is used as a masking layer. The resist layer is  $6 \mu\text{m}$  in thickness after an optical patterning. Si molds with hole diameters ranges from  $10 \mu\text{m}$  to  $50 \mu\text{m}$ , and depths exceeding  $100 \mu\text{m}$  were obtained. The micro-loading



a) Si mold fabrication by deep RIE.



b) PZT slurry casting, solidification and calcination.



c) Hot isostatic pressing (HIP)



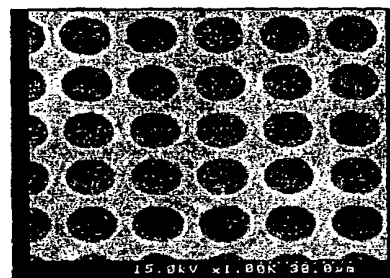
d) Removal of the Si mold by  $\text{XeF}_2$  etching.

Fig. 3 Si mold process for PZT 1-3 structure fabrication.

effect was found: the smaller the hole diameter, the shallower the etched depth. The sidewalls, however, are satisfactorily vertical and smooth for all holes. Fig. 4 a) shows the top view (tilted) of a Si mold with the hole diameter  $D = 16 \mu\text{m}$  and periodicity  $p = 22 \mu\text{m}$ . Fig. 4 b) shows the cross sectional view of a Si mold with  $D = 16 \mu\text{m}$  and  $p = 32 \mu\text{m}$ . The holes are as deep as  $140 \mu\text{m}$ . The bad cleanliness of the cross section is due to the damage during Si cleavage.

## 2) PZT Slurry Casting

The slurry was prepared by mixing PZT powder



a) top (tilted)



b) cross section

Fig. 4 SEM images of Si molds fabricated by deep RIE.

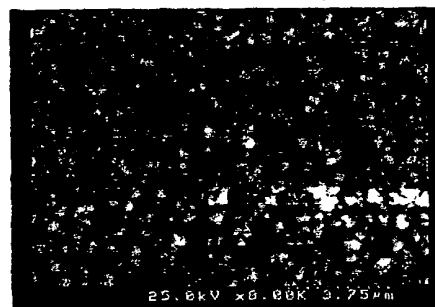


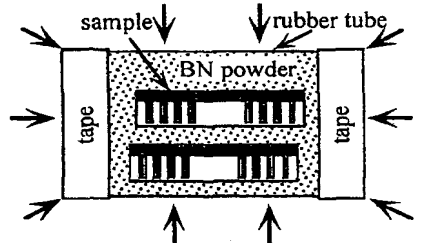
Fig. 5 PZT after slurry casting and calcination.

$(\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3)$  thoroughly with 10% PVA (polyvinyl alcohol) in a proper composition ratio. The average particle size of the PZT powder is  $0.3 \mu\text{m}$ . The PZT slurry was then cast into Si molds with the help of ultrasonic agitation. After a natural solidification for more than 12 hrs, the PZT green was calcinated at  $500^\circ\text{C}$  in the air for more than two hours (Fig. 3 b).

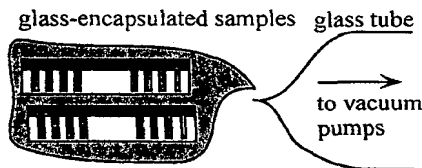
The main purpose of the calcination is to burn out the binder, PVA. At this stage, PZT has not been completely filled into the Si mold and as shown in Fig. 5, the PZT density is still low.

### 3) Hot Isostatic Pressing

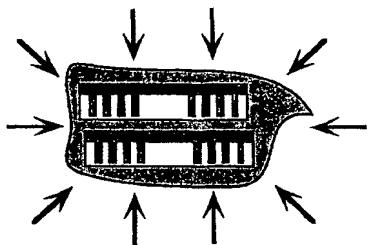
In order to condense PZT and fill it completely into the Si mold, PZT sintering was performed by glass-encapsulated hot isostatic pressing (HIP) (Fig. 3 c). The procedure of is depicted in Fig. 6.



a) Cold isostatic pressing (CIP) in water.



b) Glass encapsulation.



c) Hot isostatic pressing (HIP) in Ar gas.

Fig. 6 Procedure of hot isostatic pressing.

Initially, samples (calcinated PZT with the Si mold unremoved) were embedded into boron nitride (BN) powder and cold-isostatically pressed (CIP) at 100 MPa in water. At this stage, PZT has been compressed to a relatively high density. Then the BN-embedded samples

were sealed in a Pyrex glass tube by heating the glass tube up to its softening temperature ( $\sim 750^\circ\text{C}$ ), after having evacuated the tube inside down to  $10^{-3}$  Pa. A gas-flame burner was used to help the glass capsule envelope the samples closely and then to cut the glass capsule off the glass tube. Here BN powder was used to prevent any reaction between the samples and glass in the encapsulating and the later HIP processes. Lastly, the glass-encapsulated samples were hot-isostatically pressed with Ar gas as a pressure-transmitting medium. They were firstly heated up to the Pyrex softening temperature under a low pressure about 1 MPa. Pressure and temperature were then concurrently increased to the predetermined peak values of 70 MPa and  $1000^\circ\text{C}$ , and held for two hours. The heating and pressing program is depicted by Fig. 7.

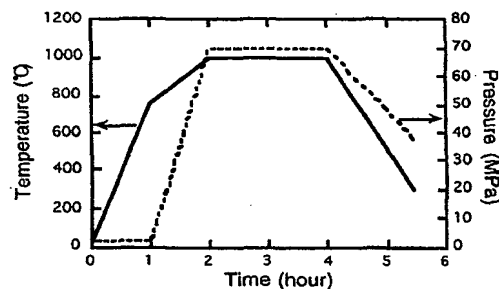


Fig. 7 Heating and pressurization program of HIP.

### 4) Si Mold Removal

After HIP, samples were taken out from the glass and BN package and were polished from the Si side until clean Si molds were exposed. Then,  $\text{XeF}_2$  etching [9] is performed to remove the Si molds (Fig. 3 d). According to an in-situ monitoring of the etching products by Fourier Transform Infrared (FT-IR) spectroscopy, there was no observable etching of PZT. The setup of our  $\text{XeF}_2$  etcher and other details can be found in Ref. 10.

## RESULTS AND DISCUSSION

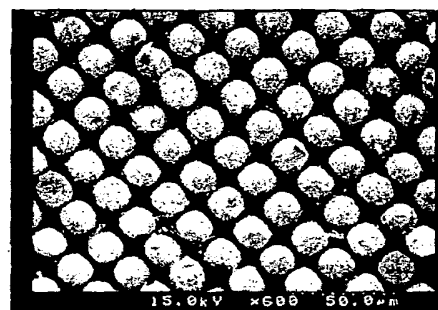
Fig. 8 shows SEM pictures of typical PZT 1-3 structures fabricated by the Si lost mold process. The displayed PZT rods are 16  $\mu\text{m}$  in diameter, 100  $\mu\text{m}$  in height, resulting in an aspect ratio higher than six. The PZT rod cross sections were also investigated by SEM observation. It is found that PZT has been highly condensed even in small PZT rods. Fig. 9 shows the cross section of a PZT rod with a diameter below 10  $\mu\text{m}$ .

The fabricated PZT structures are ready to be applied to PZT/polymer 1-3 composite transducers in the frequency range of 20 MHz. Due to the high PZT density, good transducer performance is expectable.

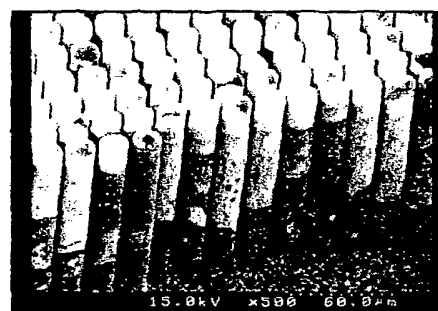
The experimental results suggest that due to the feasibility of pressing during sintering (HIP), the Si mold process has opened a new way for high density fine structure fabrication of PZT. Another notable advantage of this process is that a Si mold can be fabricated easily by Si deep RIE, which is now a quite popular technique. Such a mold can be as fine as several micrometers in diameters with aspect ratios higher than 10.

In order to remove Si molds after PZT sintering,  $\text{XeF}_2$  etching has been utilized. It may be a meaningful try to remove the Si mold by a conventional wet etching, using, for example, tetramethyl ammonium hydroxide (TMAH). This may be possible if a protection film (silicon nitride, silicon oxide, etc.) is formed on the surface of the Si mold before PZT casting. If this is possible, mass production of fine scale PZT structures at low cost will be realized.

Here, it shall be emphasized that the above process has not been optimized. For example, HIP conditions (temperature, pressure and time) are to be carefully scanned



a) top



b) side

Fig. 8 PZT rod array fabricated by Si mold process.

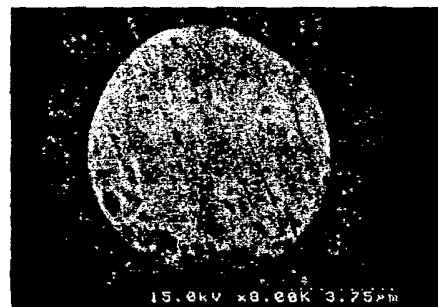


Fig. 9 SEM image of the cross section of a PZT rod.

in order to obtain high quality PZT structures (concerned factors: crystallization state, density and grain sizes, etc.), without destroying the Si mold. Slight reaction (or diffusion) seems to have occurred between Si and PZT at the interface, which is also expected to be prevented by a protection film on the Si mold surface.

## CONCLUSION

It has been demonstrated that the Si lost mold process developed in this work is a promising technique for the fabrication of fine scale high aspect ratio PZT 1-3 structures. Due to the high strength and high melting point of Si, PZT sintering under high pressures (HIP) without removing the Si mold becomes possible. This leads to structures of highly condensed PZT. The fabricated structures are suitable for PZT/polymer 1-3 composite micro-transducers in the frequency range of 20 MHz with expectable good performance.

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